2008 ARCPAC CCN Measurements Analysis of Instrument Uncertainty Richard Moore and Athanasios Nenes

1 Summary

This document discusses the estimated uncertainty associated with the reported values of CCN Number Concentration (cm⁻³) for the 2008 NOAA ARCPAC campaign. It is shown that the relative uncertainty can be expressed as a function of the reported CCN number concentration at STP:

$$\varepsilon_{CCN_{STP}} \equiv \frac{\sigma_{CCN_{STP}}}{CCN_{STP}} = \sqrt{\frac{1}{(0.455)(CCN_{STP}[cm^{-3}])} + 5.06 \times 10^{-3}}$$
(1)

where CCN_{STP} and $\sigma_{CCN_{STP}}$ are the CCN number concentration and its absolute uncertainty (standard deviation), respectively. As would be expected counting statistics constitute the largest source of uncertainty for most CCN number concentrations measured during ARCPAC (Figure 1).

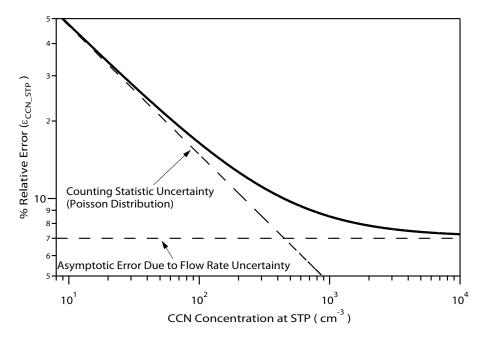


Figure 1: CCN number concentration relative uncertainty as a function of CCN number concentration at STP. The contributions to the uncertainty from counting statistics and the combined uncertainties of flow rate, temperature, and pressure are also shown.

2 CCN Number Concentration Uncertainty

The CCN number concentration is reported with 1-second resolution at Standard Temperature and Pressure (STP; 1013 mbar, 273.15 K), while measurements were made at a fixed pressure P = 450 mb and the instrument operating temperature (T[K]). The reported concentration is calculated as:

$$CCN_{STP}[cm^{-3}] = \left(\frac{N[s^{-1}]}{Q_a[cm^3s^{-1}]}\right) \left(\frac{T[K]}{273.15[K]}\right) \left(\frac{1013[mbar]}{P[mbar]}\right)$$
(2)

where $N[s^{-1}]$ is the particle counting rate per second and $Q_a[cm^3s^{-1}]$ is the instrument aerosol flow rate. The relative uncertainty associated with CCN_{STP} is determined from the relative uncertainties associated with each measurement as follows:

$$\varepsilon_{CCN_{STP}} \equiv \frac{\sigma_{CCN_{STP}}}{CCN_{STP}} = \sqrt{\varepsilon_N^2 + \varepsilon_{Q_a}^2 + \varepsilon_T^2 + \varepsilon_P^2}$$
(3)

where $\sigma_{CCN_{STP}}$ is the absolute uncertainty of CCN_{STP} and ε_i is the relative uncertainty of measurement i = N, Q, T, and P.

2.1 Uncertainty of Count Rate, ε_N

Ammonium sulfate calibration aerosol, sized using the NOAA DMA, were used to assess the uncertainty associated with the count rate during two instrument intercomparison periods on 9 April 2008 and 17 April 2008. Instrument supersaturations examined were 0.16% and 0.39% on April 9th and 0.44% on April 17th. Particle sizes in the range of 80 nm to 600 nm were sampled in a stepwise manner. During each step, concentration was allowed to stabilized before calculating the average (\bar{x}_{CCN}) and standard deviation (σ_{CCN}) of CCN concentrations at STP (approximately 100-200 points per averaging interval). The data are shown in Table 2.1.

Equation 2 can be rearranged to solve for N as follows:

$$N[s^{-1}] = \left(CCN_{STP}[cm^{-3}]\right) \left(Q_{aerosol}[cm^{3}s^{-1}]\right) \left(\frac{273.15[K]}{T[K]}\right) \left(\frac{P[mbar]}{1013[mbar]}\right) \tag{4}$$

During the campaign, the CCN counter was operated at a nearly constant total flow rate of 0.75 L min⁻¹ and a sheath:aerosol ratio of 10:1, which corresponds to an average value of Q_a of 1.136 cm³ s⁻¹. Average operating temperature and pressure were 303 K and 450 mb, respectively. Consequently, from Equation 4, $N[s^{-1}] \simeq (0.455)(CCN_{STP}[cm^{-3}])$ during the ARCPAC campaign.

This correction was applied to the calibration data to yield the values of \overline{x}_{counts} and σ_{counts} shown in Table 2.1. The experimentally-determined uncertainty, $\varepsilon_N|_{measured}$ is then just the ratio of σ_{counts} to \overline{x}_{counts} . When sampling an aerosol with constant mean number concentration (i.e., the time scale of concentration change is much greater than the sampling time interval), the count data are approximated by the Poisson distribution. This is convenient because the mean of the Poisson distribution is equal to the variance (i.e.,

Aerosol	\overline{x}_{CCN}	σ_{CCN}	\overline{x}_{counts}	σ_{counts}	Measured	Predicted
Size (nm)					Rel. Error	Rel. Error
600	8.0	4.0	3.6	1.8	50.0%	52.4%
500	27.7	7.5	12.6	3.4	27.1%	28.2%
400	55.4	11.8	25.2	5.4	21.3%	19.9%
NR	158.7	18.2	72.2	8.3	11.5%	11.8%
80	224.7	21.9	102.3	10.0	9.75%	9.89%
NR	450.0	30.0	204.8	13.7	6.67%	6.99%
225	484.9	34.1	220.7	15.5	7.03%	6.73%
180	590.8	36.4	268.9	16.6	6.16%	6.10%
NR	618.2	37.5	281.3	17.1	6.07%	5.96%
NR	1115	47	507.8	21.3	4.19%	4.44%

NR = not recorded

Table 1: Comparison between the predicted relative uncertainty and the measured relative uncertainty determined from the mean and standard deviation of concentrations of monodisperse (NH₄)₂SO₄ calibration aerosol. Predicted uncertainty calculated assuming counts are distributed as in a Poisson distribution (i.e., $\sigma_{counts} = \sqrt{\overline{x}_{counts}}$).

 $\overline{x}_{counts} = \sigma_{counts}^2$) and $\varepsilon_N|_{predicted} = \sigma_{counts}/\overline{x}_{counts} = 1/\sqrt{\overline{x}_{counts}}$. The measured and predicted relative errors are shown in Table 2.1 and in Figure 2, and it is clear that the Poisson distribution does a good job of approximating the calibration data uncertainties. Consequently, the relative uncertainty of the count rate can be expressed as

$$\varepsilon_N \simeq \frac{1}{\sqrt{(0.455)(CCN_{STP}[cm^{-3}])}} \tag{5}$$

2.2 Uncertainty of Flow Rate, ε_{Q_a}

CCN counter sheath (Q_s) and sample aerosol (Q_a) flow rates are measured by pressure transducers calibrated independently using a Drycal flow meter. The total flow rate $(Q = Q_s + Q_a)$ is actively controlled to maintain a constant total flow rate of 0.75 l min^{-1} . While the flow calibration was performed at ~ 990 mb, the operating pressure of the instrument was 450 mb for the duration of the ARCPAC campaign. Consequently, the quality of the flow calibration at multiple pressures was assessed on 14 April 2008 by inserting the Drycal flow meter just upstream of the CCN counter inlet and downstream of the critical orifice in the pressure-control unit. Measured Drycal total flow rates as a function of pressure are shown in Figure 3. For the typical total flow rate of 0.75 l min^{-1} , we use $\varepsilon_{Q_a} = 7\%$ as a conservative estimate of instrument flow rate uncertainty.

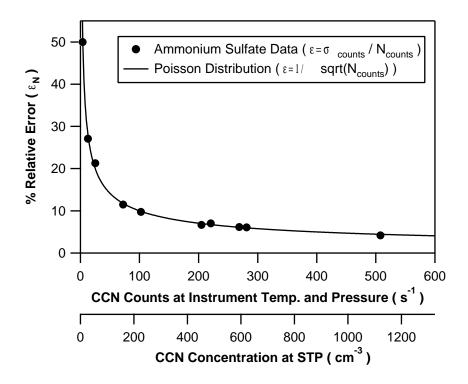


Figure 2: Relative uncertainty as a function of CCN counts per second for ammonium sulfate calibration data and assuming a Poisson distribution.

2.3 Uncertainty of Instrument Sample Temperature, ε_T

The "Sample Temperature", measured at the inlet of the CCN counter, was used to adjust the measured CCN number concentration to STP. Typical values of the sample temperature are approximately 297-303 K, and variability was observed to be fairly small ($\sim 2 \text{ K hr}^{-1}$), which is greater than the uncertainty of the RTD. Consequently, we estimate the relative uncertainty of the sample temperature to be $\varepsilon_T = (2 \text{ K})/(303 \text{ K}) = 0.6\%$.

2.4 Uncertainty of Instrument Pressure, ε_P

For all ambient pressures greater than 450 mb, the pressure in the CCN counter was actively controlled to maintain a constant pressure of 450 mb. For ambient pressures less than 450 mb, the pressure in the instrument was equal to ambient pressure. The latter case was fairly infrequent during the ARCPAC campaign, and so we estimate the uncertainty of the instrument pressure based on the 450 mb case. Variability in the pressure measurement was observed to be less than approximately ± 5 mb. Thus, we estimate the relative uncertainty of the instrument pressure to be $\varepsilon_P = (5 \text{ mb})/(450 \text{ mb}) = 1.1\%$.

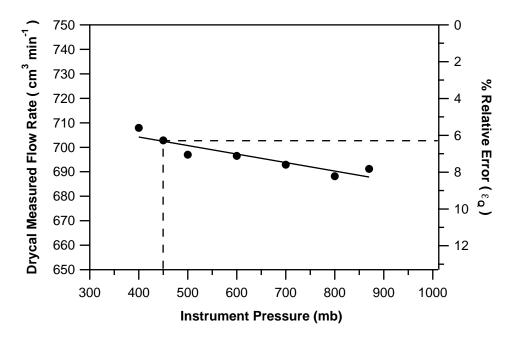


Figure 3: Drycal measured flow rate as a function of CCN counter operating pressure (left ordinate). Flow rate indicated by the CCN counter pressure transducer was approximately 750 cm³ min⁻¹ for all measurements. The percent error relative to 750 cm³ min⁻¹ is also shown (right ordinate).

2.5 Total Number Concentration Uncertainty, $\varepsilon_{CCN_{STP}}$

Using the estimated uncertainties for each measurement, we can rewrite Equation 3 using numbers:

$$\varepsilon_{CCN_{STP}} = \sqrt{\frac{1}{(0.455)(CCN_{STP}[cm^{-3}])} + 0.07^2 + 0.006^2 + 0.011^2}$$
$$= \sqrt{\frac{1}{(0.455)(CCN_{STP}[cm^{-3}])} + 5.06 \times 10^{-3}}$$